Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information for: Vibrationally Resolved Absorption Spectra and Ultrafast Exciton Dynamics in the α-Phase and β-Zinc Phthalocyanine Aggregates

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I. Supplementary Data

Table S1: Atomic coordinates (Å) of the α-phase ZnPc tetramer.

Atom	x	y	z
C	15.54458803	28.60111529	12.36491177
C	9.61985475	29.08862369	11.61375602
C	16.6010833	28.94871695	13.31182936
C	8.56335947	28.74102203	10.66683843
C	17.95709989	28.7074415	13.3219004
C	7.20734289	28.98229748	10.65676739
C	18.65254791	28.91942681	14.50735777
C	6.51189487	28.77031216	9.47131001
C	18.04395409	29.45603176	15.65783028
C	7.12048869	28.23370721	8.32083751
C	16.70603713	29.83044589	15.61246264
C	8.45840565	27.85929308	8.36620515
C	15.99048084	29.48223874	14.47258871
C	9.17396193	28.20750023	9.50607908
C	14.55399687	29.43165907	14.2267594
C	10.61044591	28.2580799	9.75190838
C	12.32685607	30.01385262	14.74841532
C	12.83758671	27.67588635	9.23025246
C	11.36195176	30.64098834	15.62553101
C	13.80249102	27.04875063	8.35313677
C	11.55752344	31.15359492	16.90673521
C	13.60691933	26.53614405	7.07193257

C	10.43375069	31.57170898	17.60518584
C	14.73069209	26.11802999	6.37348194
C	9.15133421	31.49451921	16.98305931
C	16.01310857	26.19521976	6.99560848
C	8.97787517	31.08176643	15.6614031
C	16.1865676	26.60797254	8.31726469
C	10.12050852	30.60472619	14.98136808
C	15.04393425	27.08501278	8.9972997
C	10.37510967	29.97480425	13.67304401
C	14.7893331	27.71493472	10.30562378
H	16.21448065	30.31510304	16.43435545
H	18.62140586	29.66883247	16.5544646
H	19.70143554	28.65725406	14.5410478
H	18.46804809	28.34961485	12.43859821
H	12.58856018	26.48886783	6.68261492
H	14.66997536	25.76494303	5.33491788
H	16.91973782	25.96276673	6.41774656
H	17.15755247	26.62231636	8.81005029
H	5.46300724	29.03248491	9.43761998
H	6.54303691	28.0209065	7.42420318
H	8.94996212	27.37463593	7.54431233
H	6.69639468	29.34012412	11.54006957
H	8.0068903	31.06742262	15.1686175
H	8.24470495	31.72697224	17.56092122
H	10.49446742	31.92479594	18.6437499
H	12.5758826	31.20087115	17.29605286

N	15.73746611	28.0043868	11.19134781
N	9.42697666	29.68535217	12.78731998
N	14.34579469	28.89378491	12.97157206
N	10.81864809	28.79595406	11.00709572
N	13.61895966	29.90149988	15.05282451
N	11.54548311	27.78823909	8.92584327
N	11.69950525	29.62259716	13.6065032
N	13.46493752	28.06714182	10.37216458
Zn	12.58222139	28.84486949	11.98933389
C	15.56616595	24.83187705	12.36491177
C	9.64143266	25.31938545	11.61375602
C	16.62266122	25.17947871	13.31182936
C	8.58493739	24.97178379	10.66683843
C	17.9786778	24.93820326	13.3219004
C	7.22892081	25.21305924	10.65676739
C	18.67412583	25.15018857	14.50735777
C	6.53347278	25.00107392	9.47131001
C	18.06553201	25.68679353	15.65783028
C	7.1420666	24.46446897	8.32083751
C	16.72761505	26.06120765	15.61246264
C	8.47998356	24.09005485	8.36620515
C	16.01205876	25.7130005	14.47258871
C	9.19553985	24.438262	9.50607908
C	14.57557479	25.66242083	14.2267594
C	10.63202382	24.48884167	9.75190838
C	12.34843399	26.24461439	14.74841532

C	12.85916462	23.90664811	9.23025246
C	11.38352967	26.8717501	15.62553101
C	13.82406894	23.2795124	8.35313677
C	11.57910136	27.38435668	16.90673521
C	13.62849725	22.76690581	7.07193257
C	10.45532861	27.80247075	17.60518584
C	14.75227	22.34879175	6.37348194
C	9.17291213	27.72528097	16.98305931
C	16.03468648	22.42598153	6.99560848
C	8.99945309	27.31252819	15.6614031
C	16.20814552	22.83873431	8.31726469
C	10.14208644	26.83548795	14.98136808
C	15.06551217	23.31577455	8.9972997
C	10.39668759	26.20556602	13.67304401
C	14.81091102	23.94569648	10.30562378
H	16.23605857	26.5458648	16.43435545
H	18.64298378	25.89959424	16.5544646
H	19.72301345	24.88801582	14.5410478
H	18.48962601	24.58037661	12.43859821
H	12.6101381	22.71962959	6.68261492
H	14.69155327	21.9957048	5.33491788
H	16.94131574	22.19352849	6.41774656
H	17.17913039	22.85307812	8.81005029
H	5.48458516	25.26324667	9.43761998
H	6.56461483	24.25166826	7.42420318
H	8.97154004	23.6053977	7.54431233

H	6.7179726	25.57088588	11.54006957
H	8.02846822	27.29818438	15.1686175
H	8.26628287	27.95773401	17.56092122
H	10.51604534	28.1555577	18.6437499
H	12.59746051	27.43163291	17.29605286
N	15.75904403	24.23514856	11.19134781
N	9.44855458	25.91611393	12.78731998
N	14.3673726	25.12454667	12.97157206
N	10.84022601	25.02671583	11.00709572
N	13.64053758	26.13226164	15.05282451
N	11.56706103	24.01900086	8.92584327
N	11.72108317	25.85335892	13.6065032
N	13.48651544	24.29790358	10.37216458
Zn	12.60379931	25.07563125	11.98933389
C	15.58774386	21.06263881	12.36491177
C	9.66301058	21.55014721	11.61375602
C	16.64423914	21.41024047	13.31182936
C	8.60651531	21.20254555	10.66683843
C	18.00025572	21.16896502	13.3219004
C	7.25049872	21.443821	10.65676739
C	18.69570374	21.38095034	14.50735777
C	6.5550507	21.23183569	9.47131001
C	18.08710992	21.91755529	15.65783028
C	7.16364452	20.69523074	8.32083751
C	16.74919296	22.29196941	15.61246264
C	8.50156148	20.32081661	8.36620515

C	16.03363668	21.94376227	14.47258871
C	9.21711777	20.66902376	9.50607908
C	14.5971527	21.8931826	14.2267594
C	10.65360174	20.71960343	9.75190838
C	12.3700119	22.47537615	14.74841532
C	12.88074254	20.13740988	9.23025246
C	11.40510759	23.10251187	15.62553101
C	13.84564685	19.51027416	8.35313677
C	11.60067928	23.61511845	16.90673521
C	13.65007517	18.99766758	7.07193257
C	10.47690652	24.03323251	17.60518584
C	14.77384792	18.57955352	6.37348194
C	9.19449004	23.95604274	16.98305931
C	16.0562644	18.65674329	6.99560848
C	9.02103101	23.54328996	15.6614031
C	16.22972344	19.06949607	8.31726469
C	10.16366436	23.06624972	14.98136808
C	15.08709009	19.54653631	8.9972997
C	10.41826551	22.43632778	13.67304401
C	14.83248894	20.17645824	10.30562378
H	16.25763649	22.77662657	16.43435545
H	18.6645617	22.130356	16.5544646
H	19.74459137	21.11877759	14.5410478
H	18.51120393	20.81113838	12.43859821
H	12.63171601	18.95039135	6.68261492
H	14.71313119	18.22646656	5.33491788

H	16.96289366	18.42429026	6.41774656
H	17.20070831	19.08383988	8.81005029
H	5.50616307	21.49400844	9.43761998
H	6.58619275	20.48243003	7.42420318
H	8.99311796	19.83615946	7.54431233
H	6.73955052	21.80164765	11.54006957
H	8.05004614	23.52894614	15.1686175
H	8.28786079	24.18849577	17.56092122
H	10.53762325	24.38631947	18.6437499
H	12.61903843	23.66239467	17.29605286
N	15.78062195	20.46591033	11.19134781
N	9.4701325	22.1468757	12.78731998
N	14.38895052	21.35530844	12.97157206
N	10.86180392	21.25747759	11.00709572
N	13.6621155	22.3630234	15.05282451
N	11.58863895	20.24976262	8.92584327
N	11.74266109	22.08412068	13.6065032
N	13.50809336	20.52866534	10.37216458
Zn	12.62537722	21.30639301	11.98933389
C	15.60932178	17.29340058	12.36491177
C	9.6845885	17.78090898	11.61375602
C	16.66581706	17.64100224	13.31182936
C	8.62809323	17.43330732	10.66683843
C	18.02183364	17.39972679	13.3219004
C	7.27207664	17.67458277	10.65676739
C	18.71728166	17.6117121	14.50735777

C	6.57662862	17.46259745	9.47131001
C	18.10868784	18.14831705	15.65783028
C	7.18522244	16.9259925	8.32083751
C	16.77077088	18.52273118	15.61246264
C	8.5231394	16.55157838	8.36620515
C	16.0552146	18.17452403	14.47258871
C	9.23869569	16.89978552	9.50607908
C	14.61873062	18.12394436	14.2267594
C	10.67517966	16.95036519	9.75190838
C	12.39158982	18.70613791	14.74841532
C	12.90232046	16.36817164	9.23025246
C	11.42668551	19.33327363	15.62553101
C	13.86722477	15.74103592	8.35313677
C	11.6222572	19.84588021	16.90673521
C	13.67165309	15.22842934	7.07193257
C	10.49848444	20.26399427	17.60518584
C	14.79542584	14.81031528	6.37348194
C	9.21606796	20.1868045	16.98305931
C	16.07784232	14.88750505	6.99560848
C	9.04260892	19.77405172	15.6614031
C	16.25130136	15.30025783	8.31726469
C	10.18524228	19.29701148	14.98136808
C	15.108668	15.77729807	8.9972997
C	10.43984343	18.66708955	13.67304401
C	14.85406686	16.40722001	10.30562378
H	16.27921441	19.00738833	16.43435545

H	18.68613961	18.36111776	16.5544646
H	19.76616929	17.34953935	14.5410478
H	18.53278185	17.04190014	12.43859821
H	12.65329393	15.18115312	6.68261492
H	14.73470911	14.45722832	5.33491788
H	16.98447158	14.65505202	6.41774656
H	17.22228623	15.31460165	8.81005029
H	5.52774099	17.7247702	9.43761998
H	6.60777067	16.71319179	7.42420318
H	9.01469588	16.06692122	7.54431233
H	6.76112844	18.03240941	11.54006957
H	8.07162406	19.75970791	15.1686175
H	8.30943871	20.41925753	17.56092122
H	10.55920117	20.61708123	18.6437499
H	12.64061635	19.89315644	17.29605286
N	15.80219987	16.69667209	11.19134781
N	9.49171042	18.37763746	12.78731998
N	14.41052844	17.5860702	12.97157206
N	10.88338184	17.48823935	11.00709572
N	13.68369342	18.59378517	15.05282451
N	11.61021687	16.48052438	8.92584327
N	11.76423901	18.31488245	13.6065032
N	13.52967128	16.75942711	10.37216458
Zn	12.64695514	17.53715478	11.98933389

Table S2: Atomic coordinates (Å) of the β-phase ZnPc tetramer.

Atom	x	y	z
N	17.58542678	7.17396965	6.9709536
C	17.20622825	7.7056552	8.13520617
C	18.63372317	7.62721265	6.26171678
N	17.79361216	8.71891508	8.83364545
C	15.98607676	7.26554419	8.82241864
N	19.51255061	8.6141212	6.60439846
C	19.01260407	7.05514113	4.97489393
Zn	19.40366875	9.70583333	8.25500277
C	17.03048884	8.97689613	9.96989705
C	15.06400442	6.28207151	8.51001632
C	15.86532479	8.10142026	9.9397828
C	20.47254601	8.73490059	5.60607144
C	18.46799769	6.06543731	4.17874844
C	20.1525984	7.77940983	4.56996953
N	21.01372535	10.69276129	7.6763601
N	19.29478689	10.79755517	9.90560709
N	17.3010064	9.84264676	10.95191218
C	13.99607865	6.08369398	9.40997672
H	15.1708475	5.68865686	7.59790455
C	14.79321406	7.90170333	10.83343638
N	21.50633111	9.5690005	5.55809337
C	19.10322618	5.80011865	2.96561965
H	17.59873661	5.49581166	4.47678706
C	20.75893528	7.536104	3.3287143

C	21.77684867	10.43475112	6.5401085
C	21.60110926	11.70599205	8.37479937
C	18.33479149	10.67674666	10.9039341
C	20.17361434	11.78446372	10.24828876
C	13.86004561	6.91840535	10.52060479
H	13.20860289	5.36262821	9.18920493
H	14.68256341	8.51488876	11.72573614
C	20.21759214	6.53510258	2.56245852
H	18.67660998	5.02678696	2.33276803
H	21.64427889	8.08627916	2.97912484
C	22.94201272	11.31025611	6.57022275
C	22.82126074	12.14613218	7.6875869
N	21.22191073	12.23770672	9.53905194
C	18.65473911	11.63223743	11.94003601
C	19.79473343	12.35653524	11.53511161
H	12.99893607	6.78758042	11.17938703
H	20.62192567	6.3348033	1.56002382
C	24.01412345	11.50997304	5.67656917
C	23.74333309	13.12960486	7.99998923
C	18.04840222	11.87554326	13.18129125
C	20.33933981	13.34620995	12.3312571
C	24.9472919	12.49327102	5.98940075
H	24.1247741	10.89675849	4.78426941
C	24.81125885	13.32798239	7.10002882
H	23.63649001	13.72301951	8.91210099
C	18.58974345	12.87654467	13.94755362

H	17.16306054	11.32536809	13.5308741
C	19.70411325	13.61155773	13.54437929
H	21.20860089	13.91586472	12.03321848
H	25.80840144	12.62406683	5.33061851
H	25.59873461	14.04904816	7.32080062
H	18.18541375	13.07687307	14.94997512
H	20.13072657	14.38488941	14.17724082
Zn	19.40366875	14.55875	8.25500277
N	21.01372535	15.54565854	7.6763601
N	17.79361216	13.57184146	8.83364545
N	19.51255061	13.46701845	6.60439846
N	19.29478689	15.65048154	9.90560709
C	21.77684867	15.2876775	6.5401085
C	21.60110926	16.55891843	8.37479937
C	17.03048884	13.8298225	9.96989705
C	17.20622825	12.55858157	8.13520617
C	20.47254601	13.58782696	5.60607144
C	18.63372317	12.48013903	6.26171678
C	18.33479149	15.52967304	10.9039341
C	20.17361434	16.63736097	10.24828876
C	22.94201272	16.16315337	6.57022275
N	21.50633111	14.42192687	5.55809337
C	22.82126074	16.99902944	7.6875869
N	21.22191073	17.09060398	9.53905194
C	15.86532479	12.95434663	9.9397828
N	17.3010064	14.69557313	10.95191218

C	15.98607676	12.11847056	8.82241864
N	17.58542678	12.02689602	6.9709536
C	20.1525984	12.6323362	4.56996953
C	19.01260407	11.90803839	4.97489393
C	18.65473911	16.4851638	11.94003601
C	19.79473343	17.20946161	11.53511161
C	24.01412345	16.3628703	5.67656917
C	23.74333309	17.98253124	7.99998923
C	14.79321406	12.7546297	10.83343638
C	15.06400442	11.13496876	8.51001632
C	20.75893528	12.38903037	3.3287143
C	18.46799769	10.9190625	4.17874844
C	18.04840222	16.72846963	13.18129125
C	20.33933981	18.1984375	12.3312571
C	24.9472919	17.34616827	5.98940075
H	24.1247741	15.74968487	4.78426941
C	24.81125885	18.18087965	7.10002882
H	23.63649001	18.57594589	8.91210099
C	13.86004561	11.77133172	10.52060479
H	14.68256341	13.36781513	11.72573614
C	13.99607865	10.93662035	9.40997672
H	15.1708475	10.54155411	7.59790455
C	20.21759214	11.38802895	2.56245852
H	21.64427889	12.93920553	2.97912484
C	19.10322618	10.65304502	2.96561965
H	17.59873661	10.34870891	4.47678706

C	18.58974345	17.72947105	13.94755362
H	17.16306054	16.17829447	13.5308741
C	19.70411325	18.46445498	13.54437929
H	21.20860089	18.76879109	12.03321848
H	25.80840144	17.4769932	5.33061851
H	25.59873461	18.90197453	7.32080062
H	12.99893607	11.6405068	11.17938703
H	13.20860289	10.21552546	9.18920493
H	20.62192567	11.18772967	1.56002382
H	18.67660998	9.87968422	2.33276803
H	18.18541375	17.92977033	14.94997512
H	20.13072657	19.23781578	14.17724082
N	17.58542678	16.87979328	6.9709536
C	17.20622825	17.41150794	8.13520617
C	18.63372317	17.33303628	6.26171678
N	17.79361216	18.42473871	8.83364545
C	15.98607676	16.97136781	8.82241864
N	19.51255061	18.31994483	6.60439846
C	19.01260407	16.76096476	4.97489393
Zn	19.40366875	19.41166667	8.25500277
C	17.03048884	18.68274888	9.96989705
C	15.06400442	15.98789513	8.51001632
C	15.86532479	17.80724389	9.9397828
C	20.47254601	18.44075334	5.60607144
C	18.46799769	15.77129005	4.17874844
C	20.1525984	17.48526257	4.56996953

N	21.01372535	20.39858492	7.6763601
N	19.29478689	20.5033788	9.90560709
N	17.3010064	19.5484995	10.95191218
C	13.99607865	15.78951761	9.40997672
H	15.1708475	15.39448048	7.59790455
C	14.79321406	17.60752695	10.83343638
N	21.50633111	19.27485324	5.55809337
C	19.10322618	15.50594227	2.96561965
H	17.59873661	15.20163528	4.47678706
C	20.75893528	17.24195674	3.3287143
C	21.77684867	20.14060387	6.5401085
C	21.60110926	21.4118448	8.37479937
C	18.33479149	20.38259941	10.9039341
C	20.17361434	21.49028734	10.24828876
C	13.86004561	16.62422898	10.52060479
H	13.20860289	15.06845184	9.18920493
H	14.68256341	18.2207415	11.72573614
C	20.21759214	16.24095533	2.56245852
H	18.67660998	14.73261059	2.33276803
H	21.64427889	17.7921319	2.97912484
C	22.94201272	21.01607974	6.57022275
C	22.82126074	21.85195581	7.6875869
N	21.22191073	21.94353035	9.53905194
C	18.65473911	21.33809017	11.94003601
C	19.79473343	22.06235887	11.53511161
H	12.99893607	16.49343317	11.17938703

H	20.62192567	16.04062693	1.56002382
C	24.01412345	21.21579667	5.67656917
C	23.74333309	22.83542849	7.99998923
C	18.04840222	21.581396	13.18129125
C	20.33933981	23.05206269	12.3312571
C	24.9472919	22.19909465	5.98940075
H	24.1247741	20.60261124	4.78426941
C	24.81125885	23.03380602	7.10002882
H	23.63649001	23.42884314	8.91210099
C	18.58974345	22.58239742	13.94755362
H	17.16306054	21.03122084	13.5308741
C	19.70411325	23.31738135	13.54437929
H	21.20860089	23.62168834	12.03321848
H	25.80840144	22.32991957	5.33061851
H	25.59873461	23.75487179	7.32080062
H	18.18541375	22.7826967	14.94997512
H	20.13072657	24.09071303	14.17724082
Zn	19.40366875	24.26458333	8.25500277
N	17.79361216	23.27766508	8.83364545
N	21.01372535	25.25151129	7.6763601
N	19.51255061	23.1728712	6.60439846
N	19.29478689	25.35630517	9.90560709
C	17.03048884	23.53564613	9.96989705
C	17.20622825	22.2644052	8.13520617
C	21.77684867	24.99350112	6.5401085
C	21.60110926	26.26474205	8.37479937

C	20.47254601	23.29365059	5.60607144
C	18.63372317	22.18596265	6.26171678
C	18.33479149	25.23549666	10.9039341
C	20.17361434	26.34321372	10.24828876
C	15.86532479	22.66017026	9.9397828
N	17.3010064	24.40139676	10.95191218
C	15.98607676	21.82429419	8.82241864
N	17.58542678	21.73271965	6.9709536
C	22.94201272	25.86900611	6.57022275
N	21.50633111	24.12775049	5.55809337
C	22.82126074	26.70488218	7.6875869
N	21.22191073	26.79645672	9.53905194
C	20.1525984	22.33815983	4.56996953
C	19.01260407	21.61389113	4.97489393
C	18.65473911	26.19098743	11.94003601
C	19.79473343	26.91528524	11.53511161
C	14.79321406	22.46045333	10.83343638
C	15.06400442	20.84082151	8.51001632
C	24.01412345	26.06872304	5.67656917
C	23.74333309	27.68835486	7.99998923
C	20.75893528	22.094854	3.3287143
C	18.46799769	20.62418731	4.17874844
C	18.04840222	26.43429326	13.18129125
C	20.33933981	27.90495995	12.3312571
C	13.86004561	21.47715535	10.52060479
H	14.68256341	23.07363876	11.72573614

C	13.99607865	20.64244398	9.40997672
H	15.1708475	20.24740686	7.59790455
C	24.9472919	27.05202102	5.98940075
H	24.1247741	25.45550849	4.78426941
C	24.81125885	27.88673239	7.10002882
H	23.63649001	28.28176951	8.91210099
C	20.21759214	21.09385258	2.56245852
H	21.64427889	22.64502916	2.97912484
C	19.10322618	20.35886865	2.96561965
H	17.59873661	20.05456165	4.47678706
C	18.58974345	27.43529467	13.94755362
H	17.16306054	25.88411809	13.5308741
C	19.70411325	28.17030772	13.54437929
H	21.20860089	28.47461472	12.03321848
H	12.99893607	21.34633042	11.17938703
H	13.20860289	19.92137821	9.18920493
H	25.80840144	27.18281683	5.33061851
H	25.59873461	28.60779816	7.32080062
H	20.62192567	20.8935533	1.56002382
H	18.67660998	19.58553696	2.33276803
H	18.18541375	27.63562307	14.94997512
H	20.13072657	28.94363941	14.17724082

Table S3: Vertical excitation energies ΔE and oscillator strengths of the first 16 adiabatic excited states of the α-phase ZnPc tetramer.

State	ΔE (eV)	Oscillator Strength
S_1	1.5602	0.0000
S_2	1.6251	0.0000
S_3	1.6898	0.0523
S_4	1.7467	0.0408
S_5	1.8997	0.0000
S_6	1.9440	0.0000
S_7	2.0883	1.6547
S_8	2.1016	1.9623
S_9	2.1987	0.0000
S_{10}	2.2307	0.3640
S_{11}	2.2782	0.0000
S_{12}	2.3017	0.2710
S_{13}	2.3169	0.0000
S_{14}	2.3567	0.8085
S_{15}	2.3926	0.0000
S_{16}	2.4184	0.4645

Table S4: Vertical excitation energies ΔE and oscillator strengths of the first 16 adiabatic excited states of the β-phase ZnPc tetramer.

State	ΔE (eV)	Oscillator Strength
S_1	1.5118	0.0000
S_2	1.6416	0.0775
S_3	1.8226	0.0000
S_4	1.8457	0.0000
S_5	1.8801	0.0706
S_6	1.9380	2.9002
S_7	1.9593	0.0000
S_8	2.0807	2.4743
S_9	2.2163	0.0000
S_{10}	2.2357	0.3943
S_{11}	2.2601	0.0000
S_{12}	2.2909	0.2008
S_{13}	2.3679	0.0000
S_{14}	2.3973	0.6254
S_{15}	2.4608	0.0005
S_{16}	2.4635	0.1289

Table S5: Energies, state types, and transition dipole moments (a.u.) of the quasi-diabatic states of the α-phase ZnPc tetramer.

State	Energy (eV)	Type	Transition dipole moment		
			x	y	z
$\|1,1;L_1\rangle$	1.9025	LE	3.9381	-1.0053	-0.7454
$\|1,1;L_2\rangle$	1.9615	LE	1.0256	1.5253	3.7628
$\|2,2;L_1\rangle$	1.8838	LE	3.7113	-0.9321	-0.6316
$\|2,2;L_2\rangle$	1.9438	LE	0.8657	1.4776	3.5524
$\|3,3;L_1\rangle$	1.8838	LE	3.7113	-0.9321	-0.6316
$\|3,3;L_2\rangle$	1.9438	LE	0.8657	1.4776	3.5524
$\|4,4;L_1\rangle$	1.9025	LE	3.9381	-1.0053	-0.7454
$\|4,4;L_2\rangle$	1.9615	LE	1.0256	1.5253	3.7628
$\|1,2;L_1\rangle$	2.4372	CT	-0.0013	-0.1786	0.1263
$\|1,2;L_2\rangle$	2.5277	CT	0.1110	0.2612	-0.0272
$\|2,3;L_1\rangle$	2.3028	CT	0.0145	-0.1857	0.1443
$\|2,3;L_2\rangle$	2.3931	CT	0.1232	0.2767	-0.0319
$\|3,4;L_1\rangle$	2.1827	CT	0.0136	-0.1785	0.1472
$\|3,4;L_2\rangle$	2.2677	CT	0.1346	0.2837	-0.0385
$\|2,1;L_1\rangle$	2.1827	CT	0.0136	-0.1785	0.1472
$\|2,1;L_2\rangle$	2.2677	CT	0.1346	0.2837	-0.0385
$\|3,2;L_1\rangle$	2.3028	CT	0.0145	-0.1856	0.1443
$\|3,2;L_2\rangle$	2.3931	CT	0.1232	0.2766	-0.0319
$\|4,3;L_1\rangle$	2.4372	CT	-0.0013	-0.1786	0.1262
$\|4,3;L_2\rangle$	2.5277	CT	0.1110	0.2612	-0.0272
$\|1,3;L_1\rangle$	3.3282	CS	-0.0124	0.0182	0.0156
$\|1,3;L_2\rangle$	3.4103	CS	0.0068	-0.0238	-0.0206
$\|2,4;L_1\rangle$	3.0754	CS	-0.0056	0.0157	0.0098
$\|2,4;L_2\rangle$	3.1532	CS	0.0072	-0.0226	-0.0092
$\|3,1;L_1\rangle$	3.0754	CS	-0.0053	0.0156	0.0096
$\|3,1;L_2\rangle$	3.1532	CS	0.0073	-0.0227	-0.0093
$\|4,2;L_1\rangle$	3.3282	CS	-0.0122	0.0182	0.0156
$\|4,2;L_2\rangle$	3.4103	CS	0.0068	-0.0237	-0.0202
$\|1,4;L_1\rangle$	3.6651	CS	0.0016	-0.0014	-0.0031
$\|1,4;L_2\rangle$	3.7418	CS	0.0013	-0.0016	-0.0020
$\|4,1;L_1\rangle$	3.6651	CS	0.0018	-0.0013	-0.0029
$\|4,1;L_2\rangle$	3.7418	CS	0.0016	-0.0017	-0.0021

Table S6: Energies, state types, and transition dipole moments (a.u.) of the quasi-diabatic states of the β-phase ZnPc tetramer.

State	Energy (eV)	Type	Transition dipole moment		
			x	y	z
$\|1,1;L_1\rangle$	1.8179	LE	2.0872	-0.9830	-3.7565
$\|1,1;L_2\rangle$	1.9568	LE	2.8662	3.1876	0.8834
$\|2,2;L_1\rangle$	1.8208	LE	1.9300	-1.0409	-3.6119
$\|2,2;L_2\rangle$	1.9255	LE	2.7753	3.0662	0.6948
$\|3,3;L_1\rangle$	1.8210	LE	1.9233	-1.0478	-3.6129
$\|3,3;L_2\rangle$	1.9256	LE	2.7793	3.0641	0.6870
$\|4,4;L_1\rangle$	1.8179	LE	2.0874	-0.9827	-3.7566
$\|4,4;L_2\rangle$	1.9567	LE	2.8659	3.1875	0.8837
$\|1,2;L_1\rangle$	2.3801	CT	0.0924	-0.0510	-0.1102
$\|1,2;L_2\rangle$	2.6179	CT	-0.0159	0.0434	-0.0438
$\|2,3;L_1\rangle$	2.3038	CT	0.0859	-0.0532	-0.0969
$\|2,3;L_2\rangle$	2.5323	CT	-0.0368	0.0571	-0.0434
$\|3,4;L_1\rangle$	2.2348	CT	0.0823	-0.0503	-0.1012
$\|3,4;L_2\rangle$	2.4637	CT	-0.0538	0.0566	-0.0448
$\|2,1;L_1\rangle$	2.2344	CT	0.0817	-0.0504	-0.1006
$\|2,1;L_2\rangle$	2.4634	CT	-0.0544	0.0558	-0.0450
$\|3,2;L_1\rangle$	2.3044	CT	0.0864	-0.0531	-0.0975
$\|3,2;L_2\rangle$	2.5328	CT	-0.0369	0.0571	-0.0434
$\|4,3;L_1\rangle$	2.3798	CT	0.0924	-0.0512	-0.1102
$\|4,3;L_2\rangle$	2.6176	CT	-0.0158	0.0432	-0.0442
$\|1,3;L_1\rangle$	3.3466	CS	-0.0129	-0.0043	0.0255
$\|1,3;L_2\rangle$	3.5753	CS	0.0034	0.0041	0.0064
$\|2,4;L_1\rangle$	3.2086	CS	-0.0016	-0.0041	0.0133
$\|2,4;L_2\rangle$	3.4395	CS	0.0051	0.0012	0.0044
$\|3,1;L_1\rangle$	3.2090	CS	-0.0014	-0.0037	0.0135
$\|3,1;L_2\rangle$	3.4398	CS	0.0049	0.0012	0.0044
$\|4,2;L_1\rangle$	3.3467	CS	-0.0127	-0.0039	0.0257
$\|4,2;L_2\rangle$	3.5754	CS	0.0035	0.0042	0.0063
$\|1,4;L_1\rangle$	3.7342	CS	0.0012	0.0000	-0.0011
$\|1,4;L_2\rangle$	3.9712	CS	-0.0001	-0.0018	-0.0010
$\|4,1;L_1\rangle$	3.7342	CS	0.0008	-0.0001	-0.0010
$\|4,1;L_2\rangle$	3.9710	CS	-0.0000	-0.0018	-0.0011

Table S7: Electronic couplings (meV) between the quasi-diabatic states of the α-phase ZnPc tetramer. Couplings smaller than 10 meV are not listed. The subscript of $V_{nm,n'm';L_iL_{i'}}$ (see Eq. (2) in the main text for the notation convention) is simplified for the sake of clarity.

L_1L_1			L_2L_2			L_1L_2		L_2L_1	
$V_{11,22}$ 186	$V_{11,33}$ 50.8	$V_{11,44}$ 20.0	$V_{11,22}$ 171	$V_{11,33}$ 47.6	$V_{11,44}$ 18.9				
$V_{22,33}$ 181	$V_{22,44}$ 50.8		$V_{22,33}$ 166	$V_{22,44}$ 47.6					
$V_{33,44}$ 186			$V_{33,44}$ 171						
$V_{11,12}$ -15.0	$V_{11,13}$ -17.5		$V_{11,12}$ 24.0	$V_{11,13}$ -12.1		$V_{11,12}$ -136		$V_{11,12}$ -133	
$V_{22,21}$ -20.2	$V_{22,23}$ -16.4	$V_{22,24}$ -17.8	$V_{22,21}$ 29.0	$V_{22,23}$ 27.8	$V_{22,24}$ -12.3	$V_{22,21}$ -136	$V_{22,23}$ -137	$V_{22,21}$ -132	$V_{22,23}$ -134
$V_{33,31}$ -17.8	$V_{33,32}$ -16.3	$V_{33,34}$ -20.2	$V_{33,31}$ -12.3	$V_{33,32}$ 27.8	$V_{33,34}$ 29.0	$V_{33,32}$ -137	$V_{33,34}$ -136	$V_{33,32}$ -134	$V_{33,34}$ -132
$V_{44,43}$ -15.0	$V_{44,42}$ -17.5		$V_{44,43}$ 24.0	$V_{44,42}$ -12.2		$V_{44,43}$ -136		$V_{44,43}$ -133	
$V_{11,21}$ -12.9	$V_{11,31}$ 21.8		$V_{11,21}$ -15.5	$V_{11,31}$ 21.3				$V_{11,21}$ 13.3	
$V_{22,12}$ -11.1	$V_{22,32}$ -13.9	$V_{22,42}$ 22.2	$V_{22,12}$ -14.9	$V_{22,32}$ -17.5	$V_{22,42}$ 21.8			$V_{22,12}$ 13.7	$V_{22,32}$ 14.9
$V_{33,13}$ 22.2	$V_{33,23}$ -13.9	$V_{33,43}$ -11.1	$V_{33,13}$ 21.9	$V_{33,23}$ -17.5	$V_{33,43}$ -14.9			$V_{33,23}$ 14.9	$V_{33,43}$ 13.7
$V_{44,34}$ -12.9	$V_{44,24}$ 21.8		$V_{44,34}$ -15.5	$V_{44,24}$ 21.3				$V_{44,34}$ 13.3	
$V_{12,13}$ -13.6	$V_{12,14}$ -13.0	$V_{13,14}$ -14.4				$V_{12,13}$ -133	$V_{13,14}$ -133	$V_{12,13}$ -133	$V_{13,14}$ -133
$V_{21,23}$ -15.0	$V_{23,24}$ -15.3		$V_{21,23}$ -10.8				$V_{23,24}$ -132		$V_{23,24}$ -132
$V_{32,31}$ -15.3	$V_{32,34}$ -15.0			$V_{32,34}$ -10.8		$V_{32,31}$ -132		$V_{32,31}$ -132	
$V_{43,42}$ -13.6	$V_{43,41}$ -12.9	$V_{42,41}$ -14.4				$V_{43,42}$ -133	$V_{42,41}$ -133	$V_{43,42}$ -133	$V_{42,41}$ -133
$V_{21,31}$ -27.1	$V_{21,41}$ 21.8	$V_{31,41}$ -24.3	$V_{21,31}$ -26.8	$V_{21,41}$ 21.7	$V_{31,41}$ -24.2				
$V_{12,32}$ 17.4	$V_{32,42}$ -23.7		$V_{12,32}$ 17.3	$V_{32,42}$ -23.9					
$V_{23,13}$ -23.7	$V_{23,43}$ 17.4		$V_{23,13}$ -23.9	$V_{23,43}$ 17.3					
$V_{34,24}$ -27.1	$V_{34,14}$ 21.8	$V_{24,14}$ -24.3	$V_{34,24}$ -26.7	$V_{34,14}$ 21.7	$V_{24,14}$ -24.2				

Table S8: Electronic couplings (meV) between the quasi-diabatic states of the β-phase ZnPc tetramer. Couplings smaller than 10 meV are not listed. The subscript of $V_{nm,n'm';L_iL_{i'}}$ (see Eq. (2) in the main text for the notation convention) is simplified for the sake of clarity.

L_1L_1			L_2L_2			L_1L_2		L_2L_1	
$V_{11,22}$ 186	$V_{11,33}$ 42.5	$V_{11,44}$ 12.9	$V_{11,22}$ 68.1			$V_{11,22}$ 37.0	$V_{11,33}$ 11.6	$V_{11,22}$ 28.4	$V_{11,33}$ 11.9
$V_{22,33}$ 177	$V_{22,44}$ 42.5		$V_{22,33}$ 66.3			$V_{22,33}$ 35.1	$V_{22,44}$ 11.9	$V_{22,33}$ 34.6	$V_{22,44}$ 11.5
$V_{33,44}$ 186			$V_{33,44}$ 68.1			$V_{33,44}$ 28.2		$V_{33,44}$ 37.4	
$V_{11,12}$ -58.1	$V_{11,13}$ -16.3		$V_{11,12}$ -53.2			$V_{11,12}$ 24.6		$V_{11,12}$ 18.2	
$V_{22,21}$ -59.3	$V_{22,23}$ -63.0	$V_{22,24}$ -15.8	$V_{22,21}$ -54.2	$V_{22,23}$ -53.5		$V_{22,21}$ 28.5	$V_{22,23}$ 28.2	$V_{22,21}$ 14.9	$V_{22,23}$ 17.2
$V_{33,31}$ -15.8	$V_{33,32}$ -63.0	$V_{33,34}$ -59.3		$V_{33,32}$ -53.5	$V_{33,34}$ -54.2	$V_{33,32}$ 28.4	$V_{33,34}$ 28.7	$V_{33,32}$ 17.0	$V_{33,34}$ 14.7
$V_{44,43}$ -58.1	$V_{44,42}$ -16.3		$V_{44,43}$ -53.1			$V_{44,43}$ 24.4		$V_{44,43}$ 18.3	
$V_{11,21}$ -63.2	$V_{11,31}$ 17.3		$V_{11,21}$ -80.4	$V_{11,31}$ 16.3					
$V_{22,12}$ -64.3	$V_{22,32}$ -61.1	$V_{22,42}$ 16.7	$V_{22,12}$ -79.4	$V_{22,32}$ -76.5	$V_{22,42}$ 16.5				
$V_{33,13}$ 16.8	$V_{33,23}$ -61.2	$V_{33,43}$ -64.3	$V_{33,13}$ 16.5	$V_{33,23}$ -76.5	$V_{33,43}$ -79.4		$V_{33,43}$ 10.1		
$V_{44,34}$ -63.2	$V_{44,24}$ 17.3		$V_{44,34}$ -80.4	$V_{44,24}$ 16.2					
$V_{12,13}$ -52.8	$V_{12,14}$ -11.7	$V_{13,14}$ -58.1	$V_{12,13}$ -55.3		$V_{13,14}$ -53.2	$V_{12,13}$ 23.1	$V_{13,14}$ 25.8	$V_{12,13}$ 26.2	$V_{13,14}$ 20.0
	$V_{23,24}$ -50.7			$V_{23,24}$ -54.3			$V_{23,24}$ 27.4		$V_{23,24}$ 22.1
$V_{32,31}$ -50.7			$V_{32,31}$ -54.4			$V_{32,31}$ 27.3		$V_{32,31}$ 22.2	
$V_{43,42}$ -52.9	$V_{43,41}$ -11.7	$V_{42,41}$ -58.1	$V_{43,42}$ -55.3		$V_{42,41}$ -53.3	$V_{43,42}$ 23.3	$V_{42,41}$ 25.7	$V_{43,42}$ 26.0	$V_{42,41}$ 20.1
$V_{21,31}$ -75.8	$V_{21,41}$ 15.3	$V_{31,41}$ -80.0	$V_{21,31}$ -75.8	$V_{21,41}$ 15.4	$V_{31,41}$ -80.5				
$V_{12,32}$ 12.0	$V_{32,42}$ -79.1		$V_{12,32}$ 12.9	$V_{32,42}$ -79.0					
$V_{23,13}$ -79.0	$V_{23,43}$ 12.0		$V_{23,13}$ -79.0	$V_{23,43}$ 13.0					
$V_{34,24}$ -75.8	$V_{34,14}$ 15.3	$V_{24,14}$ -80.0	$V_{34,24}$ -75.8	$V_{34,14}$ 15.4	$V_{24,14}$ -80.4				

II. Parameterization of the Aggregate Hamiltonian

The aggregate Hamiltonian for the spectral and dynamic simulations can be written as

$$
\begin{aligned}
\hat{H}_{el} =& \sum_{i}\sum_{nm} \widetilde{E}_{n,m;L_i} |n,m;L_i\rangle\langle n,m;L_i| \\
&+ \sum_{ij}\sum_{l=1}^{N-1} \widetilde{V}_{l,L_iL_j} \sum_{n=1}^{N-l} (|n,n;L_i\rangle\langle n+l,n+l;L_j| + \text{H.c.}) \\
&+ \sum_{ij}\sum_{l=1}^{N-1} t^{e,ex}_{l,L_iL_j} \sum_{n=1}^{N-l} (|n,n;L_i\rangle\langle n,n+l;L_j| + |n+l,n+l;L_i\rangle\langle n+l,n;L_j| + \text{H.c.}) \\
&+ \sum_{ij}\sum_{l=1}^{N-1} t^{h,ex}_{l,L_iL_j} \sum_{n=1}^{N-l} (|n,n;L_i\rangle\langle n+l,n;L_j| + |n+l,n+l;L_i\rangle\langle n,n+l;L_j| + \text{H.c.}) \\
&+ \sum_{ij}\sum_{l=1}^{N-1} t^{e,ct}_{l,L_iL_j} \sum_{n=1}^{N-l} \sum_{\substack{m\neq n \\ m\neq n+l}} (|m,n;L_i\rangle\langle m,n+l;L_j| + \text{H.c.}) \\
&+ \sum_{ij}\sum_{l=1}^{N-1} t^{h,ct}_{l,L_iL_j} \sum_{n=1}^{N-l} \sum_{\substack{m\neq n \\ m\neq n+l}} (|n,m;L_i\rangle\langle n+l,m;L_j| + \text{H.c.}).
\end{aligned}
\tag{S1}
$$

As compared with Eq. (2) in the main text, it can be found that we have already assumed that the state energies are location-dependent, whereas the electronic couplings are not. This is motivated by the diabatization results of the ZnPc tetramer presented in Fig. 2 and Fig. 3 in the main text. Likewise, the transition dipole moment operator of the aggregate is expressed as

$$
\hat{\boldsymbol{\mu}} = \sum_{ni} \widetilde{\mu}_{n,L_i} \left(|n,n;L_i\rangle\langle g| + |g\rangle\langle n,n;L_i| \right). \tag{S2}
$$

As compared with the symbol $\vec{\mu}_{n,m;L_i}$ used in Eq. (6) in the main text, we have introduced a new symbol $\widetilde{\mu}_{n,L_i}$ for the transition dipole moment of the aggregate to distinguish from that of the tetramer. The location-dependence of $\widetilde{\mu}_{n,L_i}$ is retained, and we have assumed a zero oscillator strength for the CT and CS states. Eq. (S1) and Eq. (S2) are parameterized based on the data of the ZnPc tetramer. In the following, we detailedly list the relation between the parameters of the tetramers and the aggregates for the α and β phases.

State energies in the middle of the aggregate for the α phase

$$\widetilde{E}_{n,n;L_1} = \frac{1}{2}(E_{2,2;L_1} + E_{3,3;L_1}) = 1.88 \text{ eV}$$

$$\widetilde{E}_{n,n;L_2} = \frac{1}{2}(E_{2,2;L_2} + E_{3,3;L_2}) = 1.94 \text{ eV}$$

$$\widetilde{E}_{n,n+1;L_1} = \widetilde{E}_{n+1,n;L_1} = \frac{1}{2}(E_{2,3;L_1} + E_{3,2;L_1}) = 2.30 \text{ eV}$$

$$\widetilde{E}_{n,n+1;L_2} = \widetilde{E}_{n+1,n;L_2} = \frac{1}{2}(E_{2,3;L_2} + E_{3,2;L_2}) = 2.39 \text{ eV}$$

$$\widetilde{E}_{n,n+2;L_1} = \widetilde{E}_{n+2,n;L_1} = \frac{1}{2}(E_{1,3;L_1} + E_{2,4;L_1}) = 3.21 \text{ eV}$$

$$\widetilde{E}_{n,n+2;L_2} = \widetilde{E}_{n+2,n;L_2} = \frac{1}{2}(E_{1,3;L_2} + E_{2,4;L_2}) = 3.28 \text{ eV}$$

$$\widetilde{E}_{n,n+3;L_1} = \widetilde{E}_{n+3,n;L_1} = E_{1,4;L_1} = 3.67 \text{ eV}$$

$$\widetilde{E}_{n,n+3;L_2} = \widetilde{E}_{n+3,n;L_2} = E_{1,4;L_2} = 3.74 \text{ eV}$$

$$\widetilde{E}_{n,n+l;L_1} = \widetilde{E}_{n+l,n;L_1} = 4.228 \text{ eV} - \frac{1.930 \text{ eV}}{l} \quad \text{for } l > 3$$

$$\widetilde{E}_{n,n+l;L_2} = \widetilde{E}_{n+l,n;L_2} = 4.288 \text{ eV} - \frac{1.900 \text{ eV}}{l} \quad \text{for } l > 3$$

State energies in the middle of the aggregate for the β phase

$$\widetilde{E}_{n,n;L_1} = \frac{1}{2}(E_{2,2;L_1} + E_{3,3;L_1}) = 1.82 \text{ eV}$$

$$\widetilde{E}_{n,n;L_2} = \frac{1}{2}(E_{2,2;L_2} + E_{3,3;L_2}) = 1.93 \text{ eV}$$

$$\widetilde{E}_{n,n+1;L_1} = \widetilde{E}_{n+1,n;L_1} = \frac{1}{2}(E_{2,3;L_1} + E_{3,2;L_1}) = 2.30 \text{ eV}$$

$$\widetilde{E}_{n,n+1;L_2} = \widetilde{E}_{n+1,n;L_2} = \frac{1}{2}(E_{2,3;L_2} + E_{3,2;L_2}) = 2.53 \text{ eV}$$

$$\widetilde{E}_{n,n+2;L_1} = \widetilde{E}_{n+2,n;L_1} = \frac{1}{2}(E_{1,3;L_1} + E_{2,4;L_1}) = 3.28 \text{ eV}$$

$$\widetilde{E}_{n,n+2;L_2} = \widetilde{E}_{n+2,n;L_2} = \frac{1}{2}(E_{1,3;L_2} + E_{2,4;L_2}) = 3.51 \text{ eV}$$

$$\widetilde{E}_{n,n+3;L_1} = \widetilde{E}_{n+3,n;L_1} = E_{1,4;L_1} = 3.73 \text{ eV}$$

$$\widetilde{E}_{n,n+3;L_2} = \widetilde{E}_{n+3,n;L_2} = E_{1,4;L_2} = 3.97 \text{ eV}$$

$$\widetilde{E}_{n,n+l;L_1} = \widetilde{E}_{n+l,n;L_1} = 4.387 \text{ eV} - \frac{2.096 \text{ eV}}{l} \quad \text{for } l > 3$$

$$\widetilde{E}_{n,n+l;L_2} = \widetilde{E}_{n+l,n;L_2} = 4.623 \text{ eV} - \frac{2.101 \text{ eV}}{l} \quad \text{for } l > 3$$

State energies at the boundaries of the aggregate for the α phase

$$\widetilde{E}_{1,1;L_1} = \widetilde{E}_{N,N;L_1} = \frac{1}{2}(E_{1,1;L_1} + E_{4,4;L_1}) = 1.90 \text{ eV}$$
$$\widetilde{E}_{1,1;L_2} = \widetilde{E}_{N,N;L_2} = \frac{1}{2}(E_{1,1;L_2} + E_{4,4;L_2}) = 1.96 \text{ eV}$$
$$\widetilde{E}_{1,2;L_1} = \widetilde{E}_{N,N-1;L_1} = \frac{1}{2}(E_{1,2;L_1} + E_{4,3;L_1}) = 2.44 \text{ eV}$$
$$\widetilde{E}_{1,2;L_2} = \widetilde{E}_{N,N-1;L_2} = \frac{1}{2}(E_{1,2;L_2} + E_{4,3;L_2}) = 2.53 \text{ eV}$$
$$\widetilde{E}_{2,1;L_1} = \widetilde{E}_{N-1,N;L_1} = \frac{1}{2}(E_{2,1;L_1} + E_{3,4;L_1}) = 2.18 \text{ eV}$$
$$\widetilde{E}_{2,1;L_2} = \widetilde{E}_{N-1,N;L_2} = \frac{1}{2}(E_{2,1;L_2} + E_{3,4;L_2}) = 2.27 \text{ eV}$$
$$\widetilde{E}_{1,3;L_1} = \widetilde{E}_{N,N-2;L_1} = \frac{1}{2}(E_{1,3;L_1} + E_{4,2;L_1}) = 3.33 \text{ eV}$$
$$\widetilde{E}_{1,3;L_2} = \widetilde{E}_{N,N-2;L_2} = \frac{1}{2}(E_{1,3;L_2} + E_{4,2;L_2}) = 3.41 \text{ eV}$$
$$\widetilde{E}_{3,1;L_1} = \widetilde{E}_{N-2,N;L_1} = \frac{1}{2}(E_{3,1;L_1} + E_{2,4;L_1}) = 3.08 \text{ eV}$$
$$\widetilde{E}_{3,1;L_2} = \widetilde{E}_{N-2,N;L_2} = \frac{1}{2}(E_{3,1;L_2} + E_{2,4;L_2}) = 3.15 \text{ eV}$$

State energies at the boundaries of the aggregate for the β phase

$$\widetilde{E}_{1,1;L_1} = \widetilde{E}_{N,N;L_1} = \frac{1}{2}(E_{1,1;L_1} + E_{4,4;L_1}) = 1.82 \text{ eV}$$
$$\widetilde{E}_{1,1;L_2} = \widetilde{E}_{N,N;L_2} = \frac{1}{2}(E_{1,1;L_2} + E_{4,4;L_2}) = 1.96 \text{ eV}$$
$$\widetilde{E}_{1,2;L_1} = \widetilde{E}_{N,N-1;L_1} = \frac{1}{2}(E_{1,2;L_1} + E_{4,3;L_1}) = 2.38 \text{ eV}$$
$$\widetilde{E}_{1,2;L_2} = \widetilde{E}_{N,N-1;L_2} = \frac{1}{2}(E_{1,2;L_2} + E_{4,3;L_2}) = 2.62 \text{ eV}$$
$$\widetilde{E}_{2,1;L_1} = \widetilde{E}_{N-1,N;L_1} = \frac{1}{2}(E_{2,1;L_1} + E_{3,4;L_1}) = 2.23 \text{ eV}$$
$$\widetilde{E}_{2,1;L_2} = \widetilde{E}_{N-1,N;L_2} = \frac{1}{2}(E_{2,1;L_2} + E_{3,4;L_2}) = 2.46 \text{ eV}$$
$$\widetilde{E}_{1,3;L_1} = \widetilde{E}_{N,N-2;L_1} = \frac{1}{2}(E_{1,3;L_1} + E_{4,2;L_1}) = 3.35 \text{ eV}$$
$$\widetilde{E}_{1,3;L_2} = \widetilde{E}_{N,N-2;L_2} = \frac{1}{2}(E_{1,3;L_2} + E_{4,2;L_2}) = 3.58 \text{ eV}$$
$$\widetilde{E}_{3,1;L_1} = \widetilde{E}_{N-2,N;L_1} = \frac{1}{2}(E_{3,1;L_1} + E_{2,4;L_1}) = 3.21 \text{ eV}$$
$$\widetilde{E}_{3,1;L_2} = \widetilde{E}_{N-2,N;L_2} = \frac{1}{2}(E_{3,1;L_2} + E_{2,4;L_2}) = 3.44 \text{ eV}$$

Electronic couplings for exciton transfer for the α phase

$$\widetilde{V}_{1,L_1L_1} = \frac{1}{3}(V_{11,22;L_1L_1} + V_{22,33;L_1L_1} + V_{33,44;L_1L_1}) = 184.3 \text{ meV}$$
$$\widetilde{V}_{1,L_2L_2} = \frac{1}{3}(V_{11,22;L_2L_2} + V_{22,33;L_2L_2} + V_{33,44;L_2L_2}) = 169.3 \text{ meV}$$
$$\widetilde{V}_{2,L_1L_1} = \frac{1}{2}(V_{11,33;L_1L_1} + V_{22,44;L_1L_1}) = 50.8 \text{ meV}$$
$$\widetilde{V}_{2,L_2L_2} = \frac{1}{2}(V_{11,33;L_2L_2} + V_{22,44;L_2L_2}) = 47.6 \text{ meV}$$
$$\widetilde{V}_{3,L_1L_1} = V_{11,44;L_1L_1} = 20.0 \text{ meV}$$
$$\widetilde{V}_{3,L_2L_2} = V_{11,44;L_2L_2} = 18.9 \text{ meV}$$

Electronic couplings for exciton transfer for the β phase

$$
\begin{aligned}
\widetilde{V}_{1,L_1L_1} &= \frac{1}{3}(V_{11,22;L_1L_1} + V_{22,33;L_1L_1} + V_{33,44;L_1L_1}) = 183.0 \text{ meV} \\
\widetilde{V}_{1,L_2L_2} &= \frac{1}{3}(V_{11,22;L_2L_2} + V_{22,33;L_2L_2} + V_{33,44;L_2L_2}) = 67.5 \text{ meV} \\
\widetilde{V}_{2,L_1L_1} &= \frac{1}{2}(V_{11,33;L_1L_1} + V_{22,44;L_1L_1}) = 42.5 \text{ meV} \\
\widetilde{V}_{3,L_1L_1} &= V_{11,44;L_1L_1} = 12.9 \text{ meV} \\
\widetilde{V}_{1,L_1L_2} &= \frac{1}{3}(V_{11,22;L_1L_2} + V_{22,33;L_1L_2} + V_{33,44;L_1L_2}) = 33.4 \text{ meV} \\
\widetilde{V}_{1,L_2L_1} &= \frac{1}{3}(V_{11,22;L_2L_1} + V_{22,33;L_2L_1} + V_{33,44;L_2L_1}) = 33.5 \text{ meV} \\
\widetilde{V}_{2,L_1L_2} &= \frac{1}{2}(V_{11,33;L_1L_2} + V_{22,44;L_1L_2}) = 11.8 \text{ meV} \\
\widetilde{V}_{2,L_2L_1} &= \frac{1}{2}(V_{11,33;L_2L_1} + V_{22,44;L_2L_1}) = 11.7 \text{ meV}
\end{aligned}
\tag{S3}
$$

Electronic couplings for exciton dissociation for the α phase

$$t^{e,ex}_{1,L_1L_1} = \frac{1}{3}(V_{11,12;L_1L_1} + V_{22,23;L_1L_1} + V_{33,34;L_1L_1}) = -17.2 \text{ meV}$$

$$t^{e,ex}_{1,L_2L_2} = \frac{1}{3}(V_{11,12;L_2L_2} + V_{22,23;L_2L_2} + V_{33,34;L_2L_2}) = 26.9 \text{ meV}$$

$$t^{e,ex}_{1,L_1L_2} = \frac{1}{3}(V_{11,12;L_1L_2} + V_{22,23;L_1L_2} + V_{33,34;L_1L_2}) = -136.3 \text{ meV}$$

$$t^{e,ex}_{1,L_2L_1} = \frac{1}{3}(V_{11,12;L_2L_1} + V_{22,23;L_2L_1} + V_{33,34;L_2L_1}) = -133.0 \text{ meV}$$

$$t^{e,ex}_{2,L_1L_1} = \frac{1}{2}(V_{11,13;L_1L_1} + V_{22,24;L_1L_1}) = -17.7 \text{ meV}$$

$$t^{e,ex}_{2,L_2L_2} = \frac{1}{2}(V_{11,13;L_2L_2} + V_{22,24;L_2L_2}) = -12.2 \text{ meV}$$

$$t^{h,ex}_{1,L_1L_1} = \frac{1}{3}(V_{11,21;L_1L_1} + V_{22,32;L_1L_1} + V_{33,43;L_1L_1}) = -12.6 \text{ meV}$$

$$t^{h,ex}_{1,L_2L_2} = \frac{1}{3}(V_{11,21;L_2L_2} + V_{22,32;L_2L_2} + V_{33,43;L_2L_2}) = -16.0 \text{ meV}$$

$$t^{h,ex}_{1,L_2L_1} = \frac{1}{3}(V_{11,21;L_2L_1} + V_{22,32;L_2L_1} + V_{33,43;L_2L_1}) = 14.0 \text{ meV}$$

$$t^{h,ex}_{2,L_1L_1} = \frac{1}{2}(V_{11,31;L_1L_1} + V_{22,42;L_1L_1}) = 22.0 \text{ meV}$$

$$t^{h,ex}_{2,L_2L_2} = \frac{1}{2}(V_{11,31;L_2L_2} + V_{22,42;L_2L_2}) = 21.6 \text{ meV}$$

Electronic couplings for exciton dissociation for the β phase

$$t^{e,ex}_{1,L_1L_1} = \frac{1}{3}(V_{11,12;L_1L_1} + V_{22,23;L_1L_1} + V_{33,34;L_1L_1}) = -60.1 \text{ meV}$$

$$t^{e,ex}_{1,L_2L_2} = \frac{1}{3}(V_{11,12;L_2L_2} + V_{22,23;L_2L_2} + V_{33,34;L_2L_2}) = -53.6 \text{ meV}$$

$$t^{e,ex}_{1,L_1L_2} = \frac{1}{3}(V_{11,12;L_1L_2} + V_{22,23;L_1L_2} + V_{33,34;L_1L_2}) = 27.2 \text{ meV}$$

$$t^{e,ex}_{1,L_2L_1} = \frac{1}{3}(V_{11,12;L_2L_1} + V_{22,23;L_2L_1} + V_{33,34;L_2L_1}) = 16.7 \text{ meV}$$

$$t^{e,ex}_{2,L_1L_1} = \frac{1}{2}(V_{11,13;L_1L_1} + V_{22,24;L_1L_1}) = -16.0 \text{ meV}$$

$$t^{h,ex}_{1,L_1L_1} = \frac{1}{3}(V_{11,21;L_1L_1} + V_{22,32;L_1L_1} + V_{33,43;L_1L_1}) = -62.9 \text{ meV}$$

$$t^{h,ex}_{1,L_2L_2} = \frac{1}{3}(V_{11,21;L_2L_2} + V_{22,32;L_2L_2} + V_{33,43;L_2L_2}) = -78.8 \text{ meV}$$

$$t^{h,ex}_{2,L_1L_1} = \frac{1}{2}(V_{11,31;L_1L_1} + V_{22,42;L_1L_1}) = 17.0 \text{ meV}$$

$$t^{h,ex}_{2,L_2L_2} = \frac{1}{2}(V_{11,31;L_2L_2} + V_{22,42;L_2L_2}) = 16.4 \text{ meV}$$

Electronic couplings for charge transfer for the α phase

$$t^{e,ct}_{1,L_1L_1} = \frac{1}{2}(V_{12,13;L_1L_1} + V_{13,14;L_1L_1} + V_{23,24;L_1L_1}) = -14.4 \text{ meV}$$

$$t^{e,ct}_{1,L_1L_2} = \frac{1}{3}(V_{12,13;L_1L_2} + V_{13,14;L_1L_2} + V_{23,24;L_1L_2}) = -132.7 \text{ meV}$$

$$t^{e,ct}_{1,L_2L_1} = \frac{1}{3}(V_{12,13;L_2L_1} + V_{13,14;L_2L_1} + V_{23,24;L_2L_1}) = -132.7 \text{ meV}$$

$$t^{e,ct}_{2,L_1L_1} = \frac{1}{2}(V_{12,14;L_1L_1} + V_{21,23;L_1L_1}) = -14.0 \text{ meV}$$

$$t^{e,ct}_{2,L_2L_2} = V_{21,23;L_1L_1} = -10.8 \text{ meV}$$

$$t^{h,ct}_{1,L_1L_1} = \frac{1}{3}(V_{21,31;L_1L_1} + V_{31,41;L_1L_1} + V_{32,42;L_1L_1}) = -25.0 \text{ meV}$$

$$t^{h,ct}_{1,L_2L_2} = \frac{1}{3}(V_{21,31;L_2L_2} + V_{31,41;L_2L_2} + V_{32,42;L_2L_2}) = -25.0 \text{ meV}$$

$$t^{h,ct}_{2,L_1L_1} = \frac{1}{2}(V_{21,41;L_1L_1} + V_{12,32;L_1L_1}) = 19.6 \text{ meV}$$

$$t^{h,ct}_{2,L_2L_2} = \frac{1}{2}(V_{21,41;L_2L_2} + V_{12,32;L_2L_2}) = 19.5 \text{ meV}$$

Electronic couplings for charge transfer for the β phase

$$t^{e,ct}_{1,L_1L_1} = \frac{1}{2}(V_{12,13;L_1L_1} + V_{13,14;L_1L_1} + V_{23,24;L_1L_1}) = -53.9 \text{ meV}$$
$$t^{e,ct}_{1,L_2L_2} = \frac{1}{2}(V_{12,13;L_2L_2} + V_{13,14;L_2L_2} + V_{23,24;L_2L_2}) = -54.3 \text{ meV}$$
$$t^{e,ct}_{1,L_1L_2} = \frac{1}{3}(V_{12,13;L_1L_2} + V_{13,14;L_1L_2} + V_{23,24;L_1L_2}) = 25.4 \text{ meV}$$
$$t^{e,ct}_{1,L_2L_1} = \frac{1}{3}(V_{12,13;L_2L_1} + V_{13,14;L_2L_1} + V_{23,24;L_2L_1}) = 22.8 \text{ meV}$$
$$t^{e,ct}_{2,L_1L_1} = \frac{1}{2}(V_{12,14;L_1L_1} + V_{21,23;L_1L_1}) = -10.8 \text{ meV}$$
$$t^{h,ct}_{1,L_1L_1} = \frac{1}{3}(V_{21,31;L_1L_1} + V_{31,41;L_1L_1} + V_{32,42;L_1L_1}) = -78.3 \text{ meV}$$
$$t^{h,ct}_{1,L_2L_2} = \frac{1}{3}(V_{21,31;L_2L_2} + V_{31,41;L_2L_2} + V_{32,42;L_2L_2}) = -78.4 \text{ meV}$$
$$t^{h,ct}_{2,L_1L_1} = \frac{1}{2}(V_{21,41;L_1L_1} + V_{12,32;L_1L_1}) = 13.6 \text{ meV}$$
$$t^{h,ct}_{2,L_2L_2} = \frac{1}{2}(V_{21,41;L_2L_2} + V_{12,32;L_2L_2}) = 14.2 \text{ meV}$$

Transition dipole moment in the middle of the aggregate for the α phase

$$\widetilde{\mu}_{n,L_1} = \frac{1}{2}(\vec{\mu}_{2,2;L_1} + \vec{\mu}_{3,3;L_1}) = (3.711, -0.932, -0.632)$$
$$\widetilde{\mu}_{n,L_2} = \frac{1}{2}(\vec{\mu}_{2,2;L_2} + \vec{\mu}_{3,3;L_2}) = (0.866, 1.478, 3.552)$$

Transition dipole moment in the middle of the aggregate for the β phase

$$\widetilde{\mu}_{n,L_1} = \frac{1}{2}(\vec{\mu}_{2,2;L_1} + \vec{\mu}_{3,3;L_1}) = (1.927, -1.044, -3.612)$$
$$\widetilde{\mu}_{n,L_2} = \frac{1}{2}(\vec{\mu}_{2,2;L_2} + \vec{\mu}_{3,3;L_2}) = (2.777, 3.065, 0.691)$$

Transition dipole moment at the boundaries of the aggregate for the α phase

$$\widetilde{\mu}_{1,L_1} = \widetilde{\mu}_{N,L_1} = \frac{1}{2}(\vec{\mu}_{1,1;L_1} + \vec{\mu}_{4,4;L_1}) = (3.938, -1.005, -0.745)$$
$$\widetilde{\mu}_{1,L_2} = \widetilde{\mu}_{N,L_2} = \frac{1}{2}(\vec{\mu}_{1,1;L_2} + \vec{\mu}_{4,4;L_2}) = (1.026, 1.525, 3.763)$$

Transition dipole moment at the boundaries of the aggregate for the β phase

$$\widetilde{\mu}_{1,L_1} = \widetilde{\mu}_{N,L_1} = \frac{1}{2}(\vec{\mu}_{1,1;L_1} + \vec{\mu}_{4,4;L_1}) = (2.087, -0.983, -3.757)$$
$$\widetilde{\mu}_{1,L_2} = \widetilde{\mu}_{N,L_2} = \frac{1}{2}(\vec{\mu}_{1,1;L_2} + \vec{\mu}_{4,4;L_2}) = (2.866, 3.188, 0.884)$$

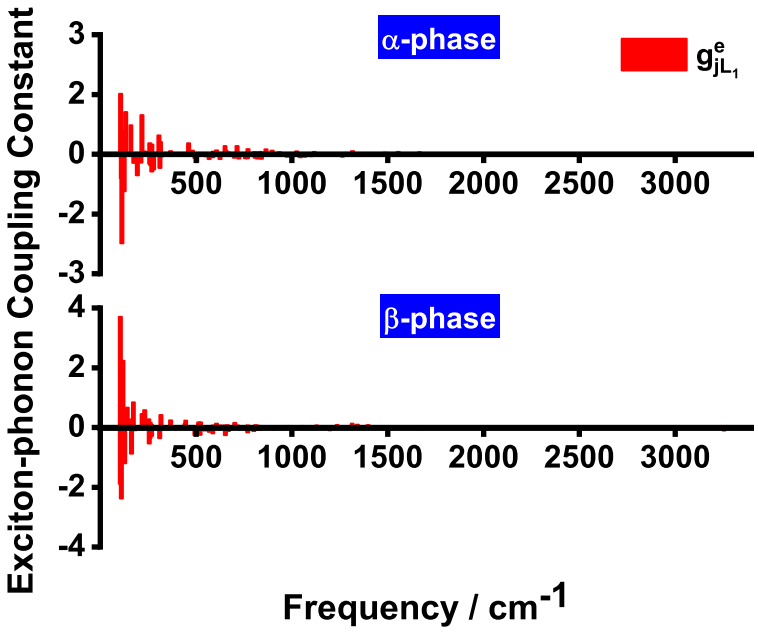

Figure S1: Electron-phonon coupling constants of all vibrational modes associated with the L_1-th localized excited states of ZnPc monomer for the α- and β-phase aggregates in the crystal environment.

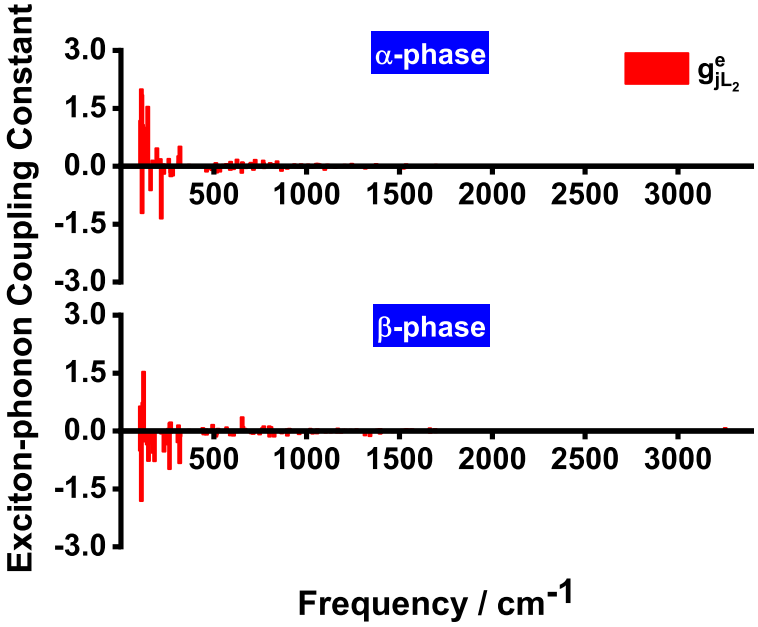

Figure S2: Electron-phonon coupling constants of all vibrational modes associated with the L_2-th localized excited states of ZnPc monomer for the α- and β-phase aggregates in the crystal environment.

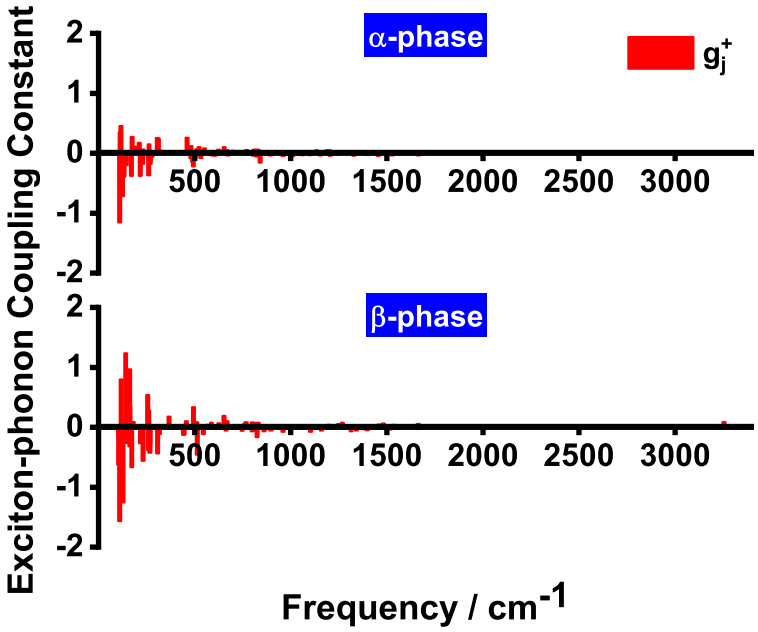

Figure S3: Electron-phonon coupling constants of all vibrational modes associated with the cationic states of ZnPc monomer for the α- and β-phase aggregates in the crystal environment.

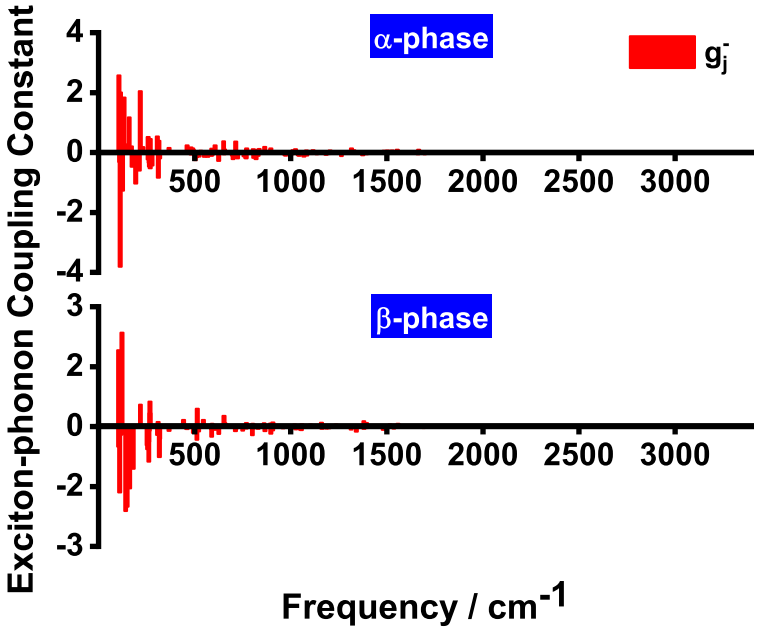

Figure S4: Electron-phonon coupling constants of all vibrational modes associated with the anionic states of ZnPc monomer for the α- and β-phase aggregates in the crystal environment.